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COVER SHEET FOR TECHNICAL MEMORANDUM

TITLE- The Influence of Atmospheric Oxygen
on Velocity of Flame Spread Along A
Solid
FILING CASE NO(S)- 320

TM- 70-2031-3

DATE- December 24, 1970

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FILING SUBJECT(S)
(ASSIGNED BY AUTHOR(S))-

ABSTRACT

The distinct, highly nonlinear variation of flamespread velocity with ambient oxygen concentration is qualitatively understandable because each of three regimes of oxygen concentration is dominated by a different rate-limiting step. These rate-limiting steps in the separate regimes of oxygen enrichment are heat transfer, pyrolysis, and gas-phase combustion.

Typically, additional oxygen stimulates a steep rise of flame speed from zero propagation, a reduced slope at moderately enriched concentrations, and another distinct rise at higher concentration. Any power law for the correlation of flame propagation velocity with oxygen partial pressure or concentration is shown to be inaccurate.

Flow diagrams illustrate the circular nature of the basic mass and energy relationships for ignition and combustion of solids.

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TECHNICAL MEMORANDUM

INTRODUCTION

The well known stimulation of flame propagation along solids by an increase of oxygen concentration in the environment has not yet been expressed accurately. There is no dependable basis for predicting the complex variation of flame spread velocity with oxygen concentration. The most popular generalizations are inadequate. It is important that this variation be predictable or at least better understood so that the flame propagation velocity associated with a material at one oxygen concentration can be reliably estimated from propagation measurements at other concentrations. This capability would permit more systematic selection of materials for use in spacecraft, submarines, test chambers, diving equipment and other inhabited systems with enriched oxygen atmospheres. It would also contribute to a more precise balance of flammability and other hazards in the selection of the oxygen concentration for such chambers.

I. A SUMMARY OF FLAME SPREAD DATA

Typical flame spread data show that there is no steady or significant propagation at oxygen concentrations just sufficient for ignitor-supported combustion. Under such conditions, the fire goes out soon after the ignition source is removed. With added oxygen, the typical flame spread rate versus oxygen curve increases sharply from zero, and, after some rather complex variations, levels off. Additional enrichment in the available oxygen stimulates another distinct increase in flame velocity. For very rapidly propagating flames in very oxygen-rich atmospheres, the curve again levels off as shown in Figs. 2, 3 and 5.

This uneven increase of flame spread velocity, as indicated by the inflections in the curves, was noted by E. M. Roth¹ and recently by A. H. Striepens.² Robinson and Gross of the National Bureau of Standards also noted a similar response of burning rate in an enclosed chamber with an increase in ventilation.³ Oxygen concentration available at the flame front in

this case was roughly proportional to the ventilation. In addition, Kimzey,⁴ and Chianta and Stoll,⁵ Cook,⁶ Tarifa,⁷ Smoot,²⁹ and their co-workers also reported similar variations. The same complex pattern of flame velocity versus oxygen concentration was reported by Vanpee for the combustion of a liquid hydrazine-diborane mixture.⁸

It is significant that these similar curves (Fig. 1-6):

- a. represent the experimental data of many dissimilar materials (paper, cotton, rubber, Teflon, Nomex, etc.)
- b. are from different sources with wide variation in specimen dimensions and ignition techniques
- c. increase in log-log nonlinearity with the number of data points within the range of oxygen concentration.

Thus, the reported highly nonlinear and distinctly patterned variation of flame propagation velocity reflects basic combustion phenomena.

II. POWER-LAW CORRELATIONS

A. Simple Power-Law Dependency

The correlation between flame spread velocity and oxygen concentration has often been approximated by a simple power-law dependency.⁹⁻¹²

$$u \sim (O_{2MF})^m \quad 1a$$

or

$$u \sim (O_{2MF}^m P)^{m'} \quad 1b$$

where u is the flame propagation velocity, O_{2MF} the mole fraction of oxygen, P the pressure, and m and m' constants of the material. This simple dependency is usually supported by a log-log plot of flame-spread-rate data.

However such log-log plots tend to mask curvature, thus giving an exaggerated impression of (logarithmic) linearity. Closer examination of the data shows that no simple power law can achieve the precise correlation claimed. Firstly, the simple power law equations presented above cannot predict the concentrations which result in zero flame propagation. Secondly, the slope of flammability versus oxygen concentration changes much too severely and unevenly for a precise representation by any simple power law.

B. An Improved Power-Law Dependency

An improvement over the simple power law dependency, which is capable of predicting zero flame spread rate, has been proposed by investigators at Atlantic Research Corporation.¹³

"The burning rate of filter paper in the upward direction at a 45° angle in the normal gravitational field is well correlated by the equation,

$$r = 4.6 \log \left[P^{0.5} / (C_p)_{O_2} + n (C_p)_x \right] + 7.8 \quad (2)$$

over the pressure range from 0.2 atm to 10 atm and for gas compositions from pure oxygen to oxygen diluted with up to 80 percent nitrogen or helium. Here r is the flame spread rate in cm./sec., P is the pressure in atmospheres, n is the number of moles of inert gas per mole of oxygen, and C_p is the heat capacity at constant pressure for oxygen or the inert gas."

The first term is analogous to the simple power law equation (1b) and the constants are functions of the material. The above equation predicts the demonstrated relative effectiveness of high versus low molar specific heats of inert diluents (N_2 versus He). Although superior to simpler forms of the power law, the fit of this equation also becomes less satisfactory as the range and the number of experimental points increase. This equation was originally based on Cook's experimental results.⁶ The plot of the data versus the prediction in Fig. 4 shows the inadequacy of this form of the power law for predicting flame propagation velocity.

The equation was also compared with typical experimental results reported by the NASA Manned Spacecraft Center on the combustion of materials considered for use in the Apollo Spacecraft. Again, the power law was shown to be inadequate for extrapolation over wide ranges of oxygen concentration.

If the constants are permitted to vary with oxygen concentration, the resulting curve can be made to fit this or any other similar data. The accuracy and usefulness of any such equations, however, is limited to the quantity and quality of experimental points for each material.

III. PRELIMINARY CONSIDERATIONS

A. General Relationships of Dominant Processes

The basic relationships of combustion assumed here are relatively well known and consistent with recent general statements by Tarifa, de Ris, Martin and their co-workers.^{7,14-16} The basic ideas are illustrated in Figs. 8 and 9 and summarized in Fig. 10. A brief study of these flow graphs will provide a sufficient basis for proceeding.

Heat transfer from the ignition source to the solid fuel starts combustion by supplying sufficient heat to initiate pyrolysis, which is the liberation at high temperatures of gases and vapors by thermal decomposition of the solid chemical structure. These pyrolysis products, when sufficiently heated, will combine with available oxygen in the combustion process, which, in turn, supplies the required heat to pyrolyze the adjacent not yet burning solid. Thus, the flame acts as the ignitor in the continuous process of gas-phase ignition required for flame propagation.^{7,10,14,16} Each of these processes, heat transfer, pyrolysis and gas-phase combustion, is sensitive to the gaseous environment and temperature. All are known to be non-linear functions of oxygen concentration.

B. Experimental Observations Related to Flame Spread Velocity

The general relationships summarized in the combustion flow charts (Figs. 8 to 10) will lead to better understanding of the physical interactions if related experimental observations are studied.

1. Ignitor Energy

Striepens (see Fig. 5), Cook, and others have noted that ignition can occur in lower concentrations of ambient oxygen with increased ignitor energy or temperatures.^{2,6,9} For instance, ignition of a resin glass-fabric laminate occurs at 40% instead of 70% oxygen when a more energetic ignitor is used.

2. Flame Size

Flame size of burning solids has been shown to increase with rate of pyrolysis by Von Elbe et al.¹⁷ The same studies show a positive but limited correlation between flame size and oxygen pressure for acrylics. Stronger positive correlations were reported by Fenimore and Martin.¹⁸

3. Pyrolysis Rates With Temperature

The Thermogravimetric Analysis (TGA) of many materials shows that the weight loss due to pyrolysis varies nonlinearly with temperature, (Figs. 11 to 14). Generally, the rate of pyrolysis increases sharply with temperature immediately above the initial decomposition temperature, experiences one or two peaks, and then drops off sharply as the temperature increases further. This is the result, of course, of the relative scarcity of residual volatile material by the time high temperatures have been reached.

4. Upward-Versus-Downward Ignition and Burning

The curves of upward burning and downward burning are different in two respects - shape and scale. The upward flame velocity seems to level off at very high concentrations (Figs. 2 and 3). The downward velocity continues to increase at a steep rate (Figs. 1 and 6). The upward burning rate is reported to be 3 to 15 times greater than that of downward or horizontal burning.^{2,12} Some flame resistant materials, for instance 10-mil Teflon and 40-mil silicon-resin laminate, would burn only if ignited at the bottom and permitted to burn upward.² This is expected because during normal positive "g" burning, upward burning is stimulated by the upward flow of hot gases that engulf and preheat the as-yet-unburned solid above the rising flame front.

5. Flame Temperature

In gas-phase combustion, flame temperature initially increases with increasing oxygen, reaches a peak and then decreases as additional oxygen contributes to a fuel-lean combustion mixture.²³

6. Burning Rate In Gas-Phase Combustion

The rate at which fuel gas burns in a fuel-rich mixture increases smoothly as the concentration of oxygen is increased, as shown in a typical curve, Fig. 15. The power of the

burning-rate-versus-oxygen concentration relationship, however, gradually decreases to zero.²⁴

C. Preliminary Analysis

On the basis of the continuity equation, the rate of steady state flame propagation;

$$u = \dot{m}_p / \rho A \quad (3)$$

where \dot{m}_p is the mass rate of pyrolysis and ρ and A are the mass density of pyrolyzing components per unit volume and cross-sectional area of the burning solid, respectively. The pyrolysis rate can be expressed as

$$\dot{m}_p = M_s Q / E_p \quad (4)$$

where Q is the net heat transfer into the pyrolyzing material, E_p the heat of pyrolysis and M_s the specie molecular weight. It has been shown by Reich and Levi²⁵ and Madorsky²⁶ that even though the pyrolysis rate itself changes, the energy required for pyrolysis is relatively constant. For instance, E_p for TFE Teflon remains close to 70 Kcal/mole throughout.

The rate of net heat transfer and therefore the rate of pyrolysis and flame propagation is sensitive to the sum of the ignitor- and flame-supplied energy ($Q_i + Q_f$) minus that energy lost from the pyrolyzing material to the environment (Q_e).

$$Q = Q_i + Q_f - Q_e \quad (5)$$

In terms of temperature differences,

$$Q = H_i (T_i - T_p) + H_f (T_f - T_p) - H_e (T_p - T_e) \quad (6)$$

where H_i , H_f and H_e are the heat transfer functions between the ignitor and the pyrolyzing material, the flame and the pyrolyzing material, and the material and the environment respectively, and

where T_i , T_p , T_f and T_e are the temperatures of the ignitor, the pyrolyzing material, the flame and the environment, respectively.

The flame temperature T_f is a sensitive function of both the local oxygen concentration and the material, while the pyrolysis temperature T_p is a function of the material. Thus, it is clear from a study of equation 6 that when materials which degrade at low temperatures are placed in highly oxygen enriched environment, they burn with a higher initial flame-to-surface ΔT , $(T_f - T_p)$, and therefore can be more easily ignited. That is, if T_f is high (because of oxygen enrichment) and T_p is low, T_i or H_i need not be very high. If the material pyrolyzes at a very low temperature, the temperature differences $(T_i - T_p)$ and $(T_f - T_p)$ may be sufficient for ignition even if T_i or T_f is relatively low.

For self-sustaining combustion the term in equation 6 representing the ignition energy will be zero. Since the same heat is required to activate the reactants, the flame must be a more effective heat source when the ignitor is not effective. The size and temperature of the flame are positive functions of oxygen concentration. Therefore, in order to reach the required level of heat transfer to support steady state pyrolysis and combustion without the assistance of an ignitor there is a critically required concentration of oxygen. This critical oxygen concentration which can just support sustained combustion without an ignitor can be called the first critical concentration, $[O_2]_1$ as noted (Fig. 16). The flame propagation rate in this condition of minimum self-sustaining combustion may be very slow, but it must be positive. A flame must travel or die.

Thus the heat flux from the flame which determines pyrolysis rate is the result of the flame propagation rate, which is, in turn, a positive function of pyrolysis. This circular flow of causation in combustion is expressed in Fig. 10.

IV. HYPOTHESIS - RATE OF FLAME TRAVEL ALONG SOLID SURFACES IS DETERMINED BY THREE RATE-LIMITING STEPS

It has been demonstrated that no simple generalization explains the peculiar, highly nonlinear variation of the speed of flame propagation along a solid with the increase of atmospheric oxygen. It is my hypothesis that each of the variations of flame speed with oxygen enrichment represents a distinct condition or regime of combustion. Further, each regime will be

qualitatively explained by a dominant rate-limiting step which determines the variation of flame speed within the limits of atmospheric oxygen concentration associated with that regime.

The three separate regimes of oxygen concentration, the heat-transfer regime, the pyrolysis regime, and the gas-phase combustion regime, are so named because the rate-limiting steps that determine the response of flame-spread velocity to oxygen concentration in each of the three regimes are heat transfer, pyrolysis and gas-phase combustion.

A. Heat Transfer

As shown in the preliminary analysis, the minimum oxygen concentration required to sustain combustion is most sensitive to heat transfer processes. The prerequisites for initial ignition are known:

1. the temperature at the solid is sufficient to initiate pyrolysis, and
2. the temperature of the fuel-oxygen mixture is sufficient to initiate combustion there.

The first state in ignition, ignitor-sustained combustion, is reached when some initial combustion occurs and is sustained by the ignitor. Energy is derived during this stage both from the ignitor and initial local combustion.

During this ignitor-sustained stage, the loss of the ignitor usually means the loss of the flame. This condition of oxygen concentration, which just barely supports combustion such that the flame extinguishes when ignition energy is removed, is usually identified as supporting combustion at zero flame-spread velocity (Fig. 16).

Heat transfer is the rate-limiting step when the oxygen concentration is just sufficient for ignition. It is reasonable to expect, therefore, that the minimum required concentration will vary greatly with differences in heat transfer. Such is clearly the case. Considerations during ignition are the energy of the ignitor, location of ignitor, and the surface properties of the solid. It has been shown that the greater the ignitor energy, the weaker the concentration of ambient oxygen required for ignition (Fig. 5). Similarly, a small initial flame at the bottom edge of a solid is much more effective in transmitting heat into the pyrolyzing material above than a like flame at the top of the solid is in transmitting heat into the material below. As a result of this heat-transfer difference, ignition at the bottom can

occur with a lower oxygen concentration than ignition at the top. Numerous findings on the ease of ignition along rough or pilose surfaces (with very intense local heat transfer characteristics) also serve to demonstrate that heat transfer is the rate-limiting step when there is just sufficient oxygen to support combustion.

B. Pyrolysis

Once sustained combustion is achieved, the rate of flame spread is limited by the rate at which combustible gaseous material is driven from the solid into the surrounding oxygen mixture. The heat transfer from the flame into the pyrolyzing solid is, as noted above, sensitive to the temperature of the flame which is, in turn, dominated by the availability of oxygen. Increased concentration of oxygen ($>[O_2]_1$) causes an increased flame temperature and size and therefore increases heat transfer into and temperature in the pyrolyzing solid. One would expect, therefore, that the increase of flame velocity with oxygen concentration would be proportional to the increase of pyrolysis with temperature. This proportionality follows from equation 3 and the above reasoning.

It is noteworthy that the increase of flame spread velocity with oxygen concentration follows very closely the pyrolysis versus temperature plot (TGA) as in Figs. 13 and 14. This is the rate versus temperature plot as in Fig. 11 integrated over the range of temperatures of the solid.

As high flame temperature is reached (as a consequence of high oxygen concentration at the flame), the supply of flammable gases from the solid into the surrounding oxygen-fuel mixture would be maximized. This leveling off of pyrolysis at the maximum rate, according to this hypothesis, is the physical cause of the leveling off of the rate of combustion with intermediate oxygen concentration (see Figs. 3, 4, 6, and 7). When the pyrolysis rate is maximized, the resultant mixture is probably fuel-rich. Beyond this point, the regime changes from one in which pyrolysis is the rate-limiting step to the final one, wherein gas-phase combustion is the rate-limiting step.

C. Gas-Phase Combustion

The final rate-limiting step is gas-phase combustion. When pyrolysis has been maximized (the oxygen concentration $> [O_2]_2$), the combustion rate is limited only by the availability of oxygen, pressure and the specific heat of the total mixture. Gas-phase combustion has been thoroughly studied and the results as applied to solids clearly presented by McAlevy, his associates, and others.^{10-12,14} The characterization of self-sustaining

combustion as continuous, gas-phase ignition is useful and enlightening. And as long as our attention is limited to this final gas-phase combustion regime (after the locale is fuel saturated by high pyrolysis rates) the theoretical results of such characterization are valid.

The oxygen concentration $[O_2]_2$ at which gas-phase combustion becomes the rate-limiting step, is a sensitive function of the material (see Figs. 1-5). With resistant materials, high oxygen concentrations (as much as 80 or 90% in some cases) are required to initiate the gas-phase combustion regime. However, with paper, cotton, and other flammable materials this regime seems to begin at about 30 or 40%. Some materials (typically, solid propellants) contain chemically available oxygen. When such materials are ignited sufficient oxygen is locally available to assure flame temperatures which can sustain a very high rate of pyrolysis. Therefore, with solid propellants it can be expected that gas-phase combustion becomes the rate-limiting step at low concentrations of ambient oxygen. It is for these materials that the power law for flame-spread velocity is most accurate.

However, as noted in Observation No. 6 and shown in Fig. 15 on gas-phase combustion, the power law is valid only for a limited range of oxygen enrichment. Beyond that, the combustion rate levels off. The similarity between the gas-phase combustion curve and the region of highest propagation rates along solid samples which are either highly flammable or burning upward within an enriched atmosphere supports the characterization of this region as gas-phase combustion limited.

V. SOME CONSEQUENCES AND APPLICATIONS OF THE HYPOTHESIS

Although not yet substantiated by experimentation, the following are indicated:

1. A flame temperature plateau will be observed just prior to the beginning of the gas-phase combustion regime. This is a result of the fuel-rich combustion mixture which can be expected to occur at maximum pyrolysis rate.
2. For each material and configuration (size, surface, position, etc.) a maximum flame spread velocity will be observed in the gas-phase regime. Additional oxygen enrichment will cause no further increase.

The ideas presented here may hopefully be applied toward improved material rating and flame retardation.

1. A flammability rating system can be developed by the use of the two critical inflection points i.e., the concentration of oxygen required to just sustain minimum steady-state combustion $[O_2]_1$ (as proposed by Fenimore and Martin)¹⁸ and the concentration required to initiate gas-phase combustion, $[O_2]_2$ (Fig. 16).

In other words, the movement of the flame-velocity versus O_2 curve, Fig. 16, to the right can be useful as a measurement of improved flame resistance.

2. The search for more effective flame retardent additives and configurations may be stimulated by the study of the flow charts, Figs. 8 and 9 and the causation flow chart Fig. 10 as well as the related analysis.

CONCLUSIONS

It is clear from the data of solid fuels burning in oxygen-bearing atmospheres that: (1) the flame spread velocity increases in a distinct and highly nonlinear (and non-power-law) manner, (2) typically, the flame velocity increases dramatically as oxygen content goes up just above that minimum required for the support of combustion; levels off in a manner which is similar to pyrolysis rates at high temperature; and then increases further with additional oxygen enrichment in a manner which is similar to the increase of combustion rates of fuel gas as a function of oxygen concentration.

The distinct, highly nonlinear variation of flame-spread velocity with ambient oxygen concentration is understandable because each of three regimes is dominated by a different rate-limiting step; first, heat transfer at the minimum combustion-supporting oxygen concentration; then, pyrolysis with more oxygen than is required to just support self-perpetuating combustion; and finally, gas-phase combustion in the most oxygen enriched atmospheres. This preliminary hypothesis is consistent with available related data.

In order that this approach be verified a mathematical model of the process (as suggested by the flow charts) must be constructed. To construct and validate such a model information is needed on:

1. The local temperatures and heat transfer between the flame (and/or ignitor) and the pyrolyzing solid as a function of the ambient gas mixture.

2. Pyrolysis (products and mass-loss rate) as a function of temperature, heating rate, and ambient oxygen.
3. Effects of molecular transport variables on gas-phase combustion.
4. The more detailed variation of flame spread velocity versus oxygen concentration; typically, about 30 points between initial zero velocity and 100% oxygen should be sufficient.

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ACKNOWLEDGEMENTS

The work was performed for Manned Space Flight, National Aeronautics and Space Administration under Contract NASW-417. Earlier work was previously reported.^{27,28}

The encouragement, assistance, and valuable advice given by J. A. Saxton are acknowledged with appreciation.

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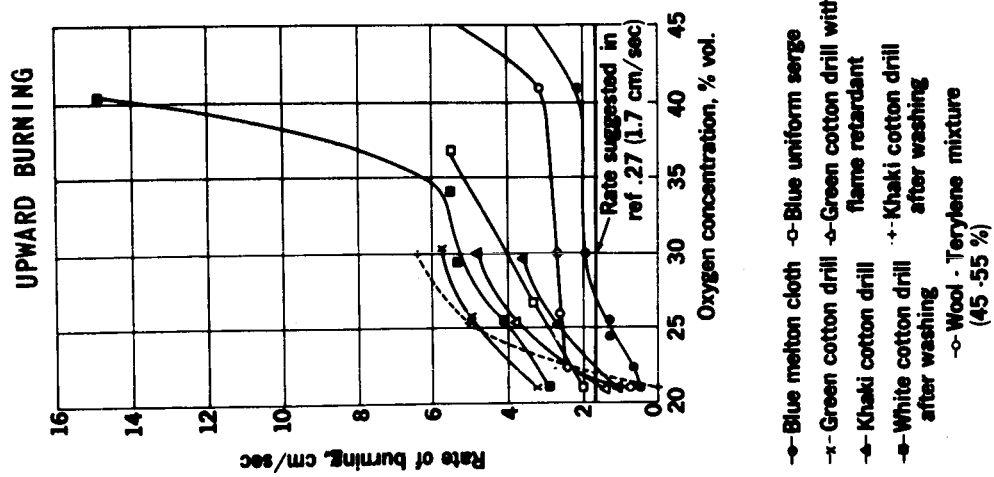


Fig. 1 - Variation of Burning Rate with Oxygen Concentration.
 (From Roth¹, Fig. 20, p. 26).

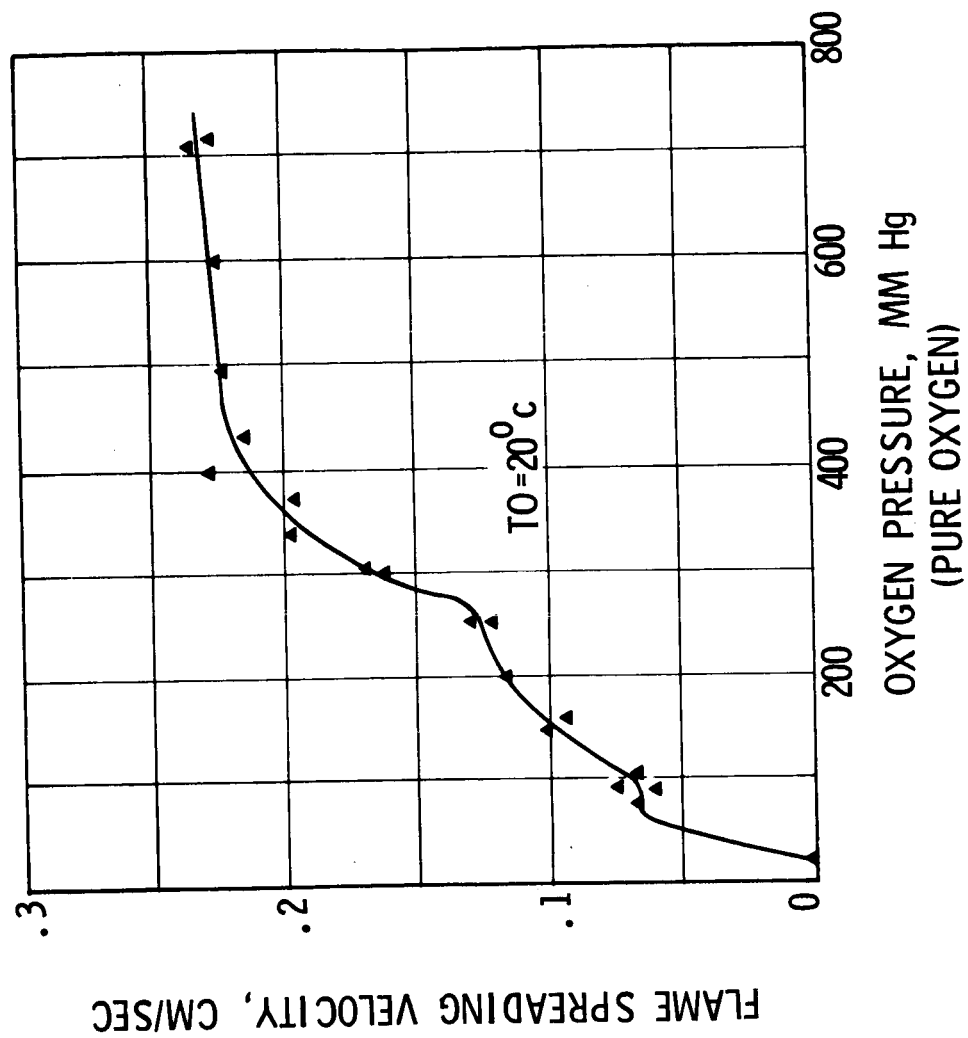


Fig. 2 - Flame Spreading Velocity Over Plexiglass Rods in Pure Oxygen. (From Tarifa¹⁴, Fig. 6A).

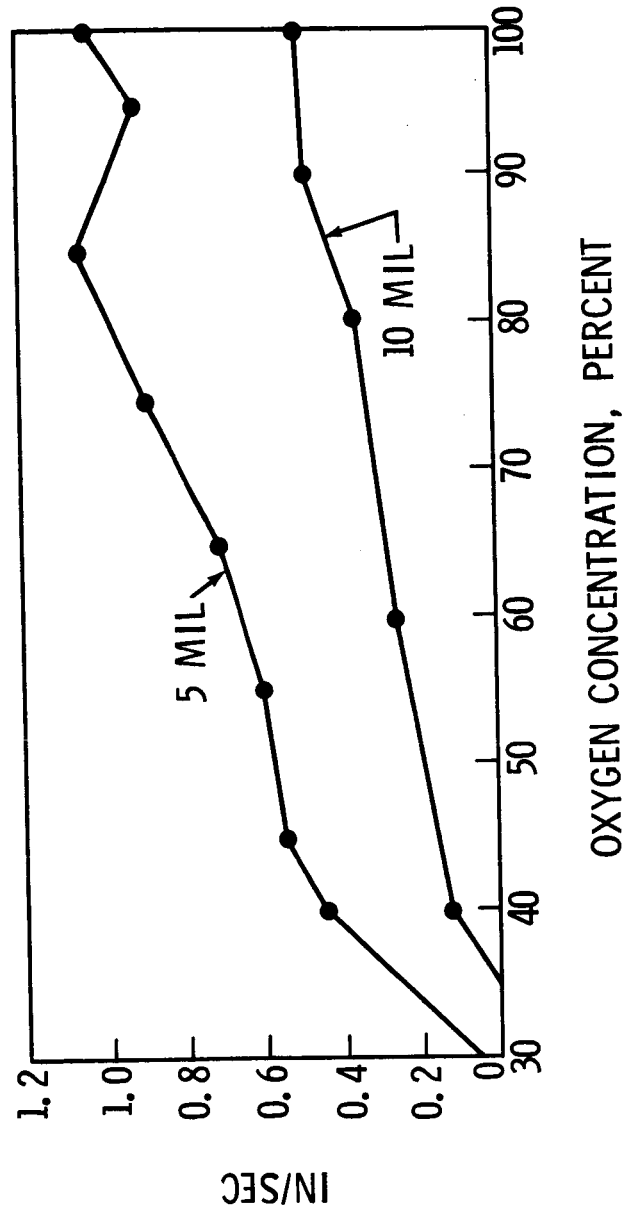


Fig. 3 - Upward Propagation of Teflon in Various O_2/N_2 Mixtures at a Total Pressure of 16.5 PSIA. (From Striepens² and Kimzey⁴).

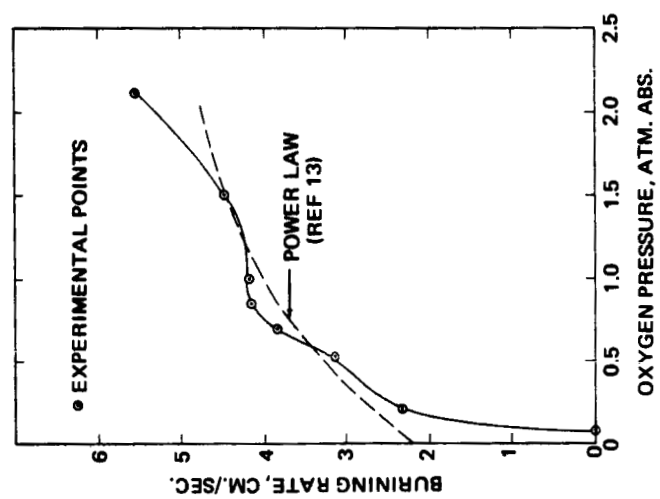


Fig. 4 - Burning Rate of Filter Paper at 45° Angle. (From Cook⁶, Fig. 34).

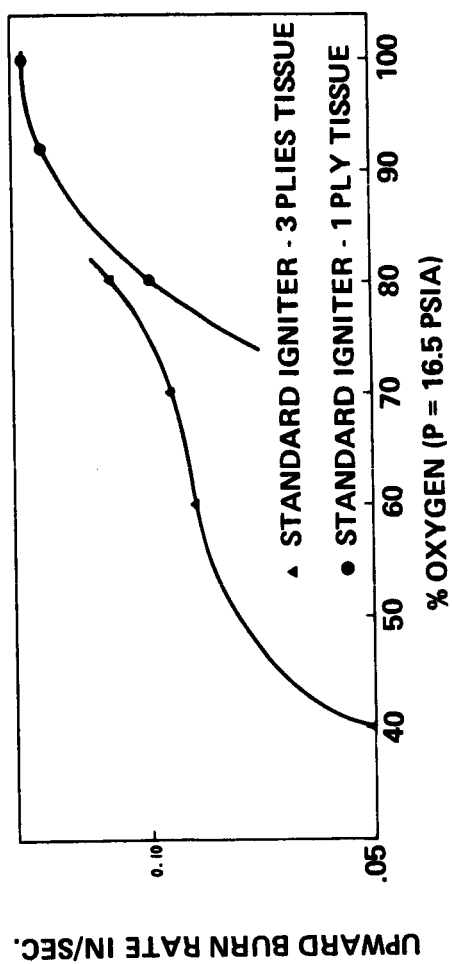


Fig. 5 - Combustion Rate of DC 7141 Silicone Resin/181 Glass Fabric Laminate (0.040 in. Thick). From Striepen²).

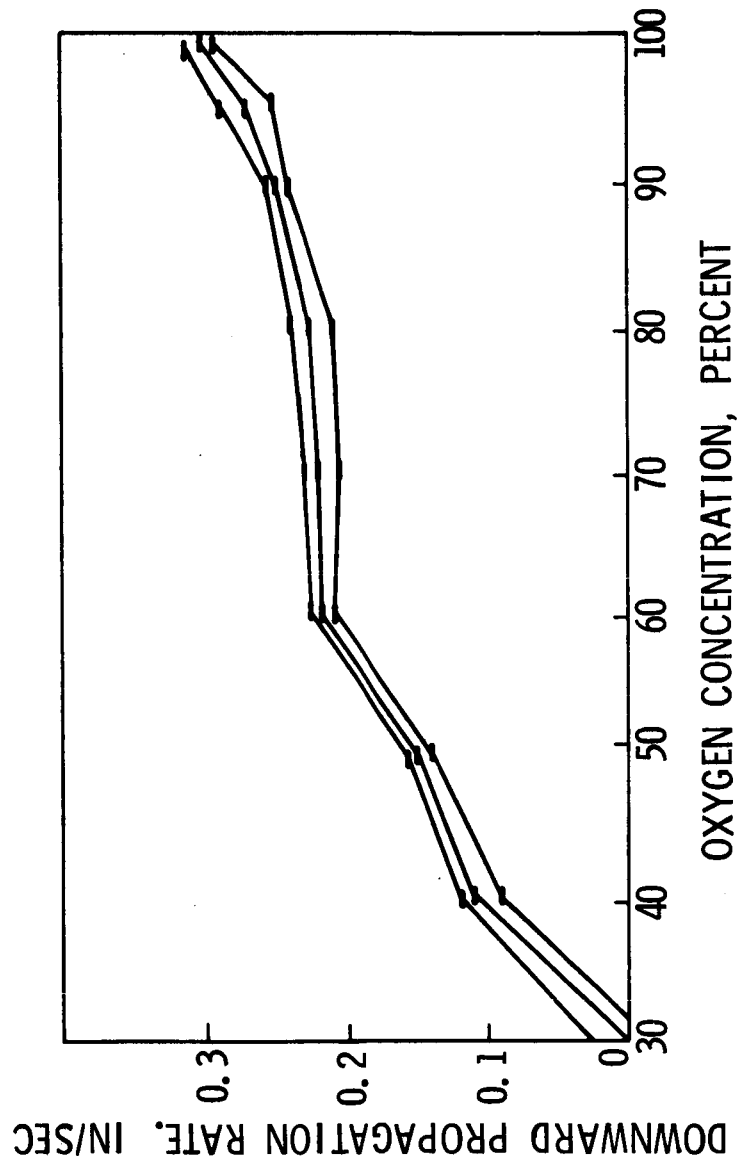


Fig. 6 - Downward Propagation of 0.005 Inch Nomex in Various O_2/N_2 Mixtures at a Total Pressure of 16.5 PSIA.
(From Kimzey⁴).

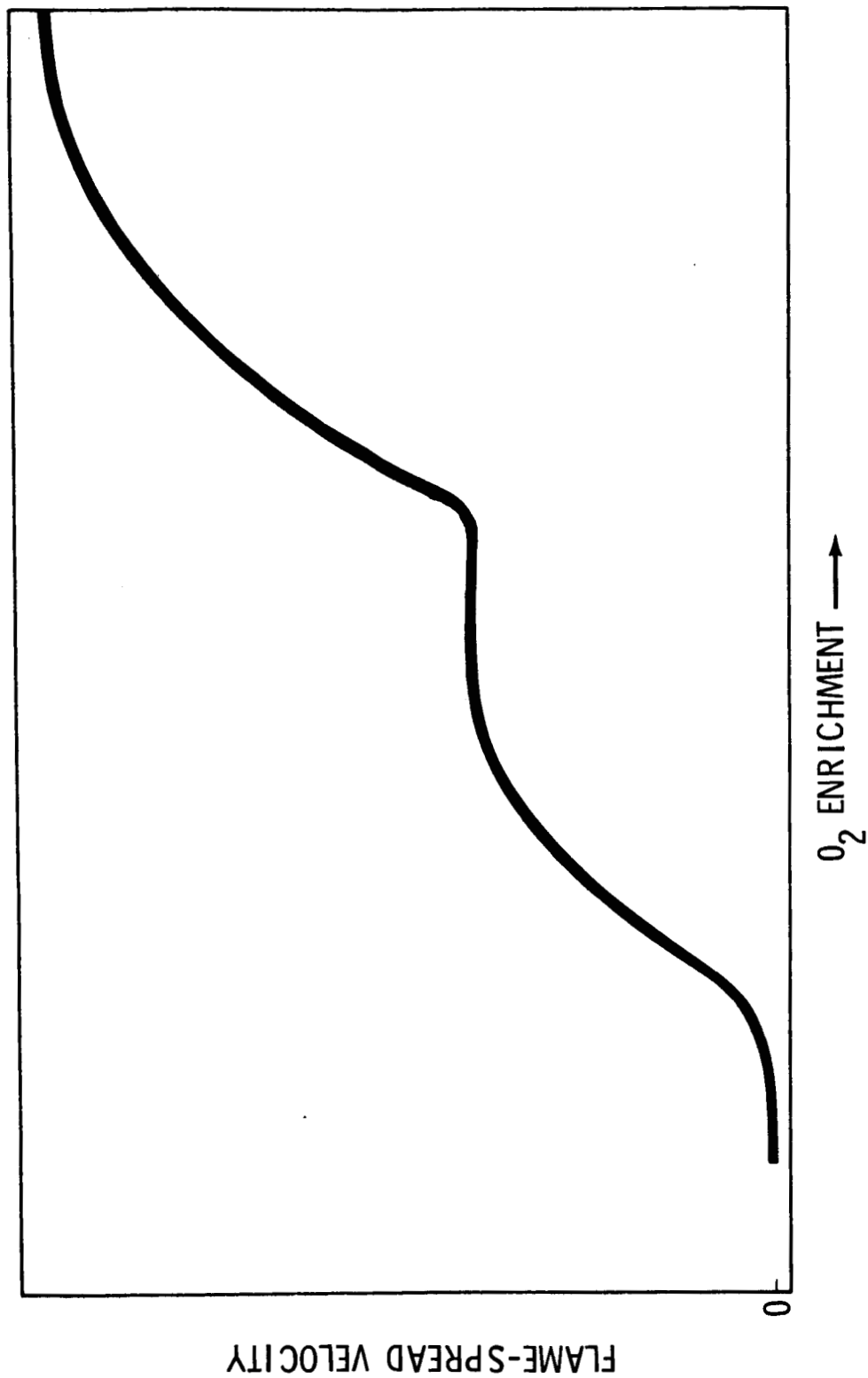


Fig. 7 - Typical Flame Spread Velocity Versus O₂ Enrichment Curve.

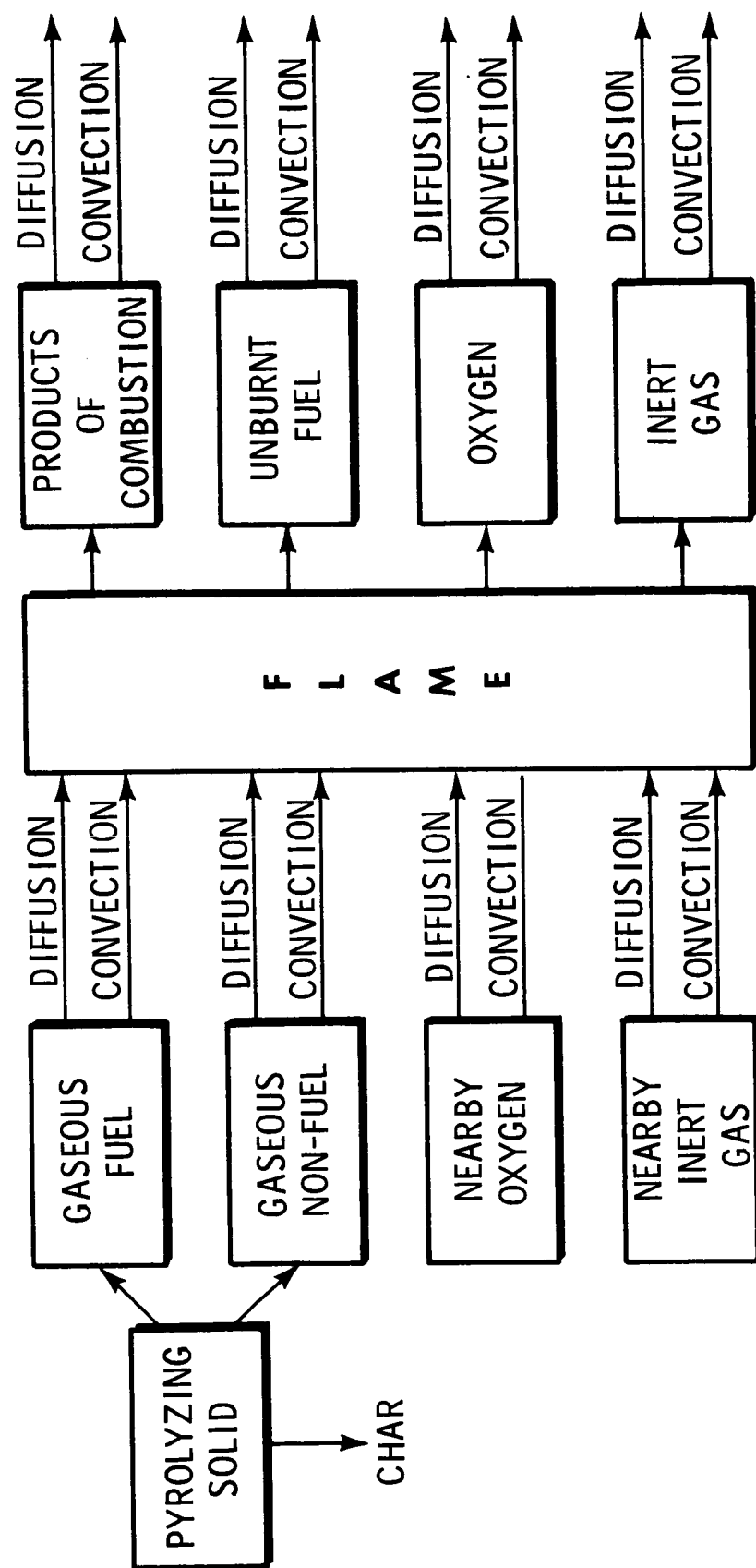


Fig. 9 - Combustion Mass Flow

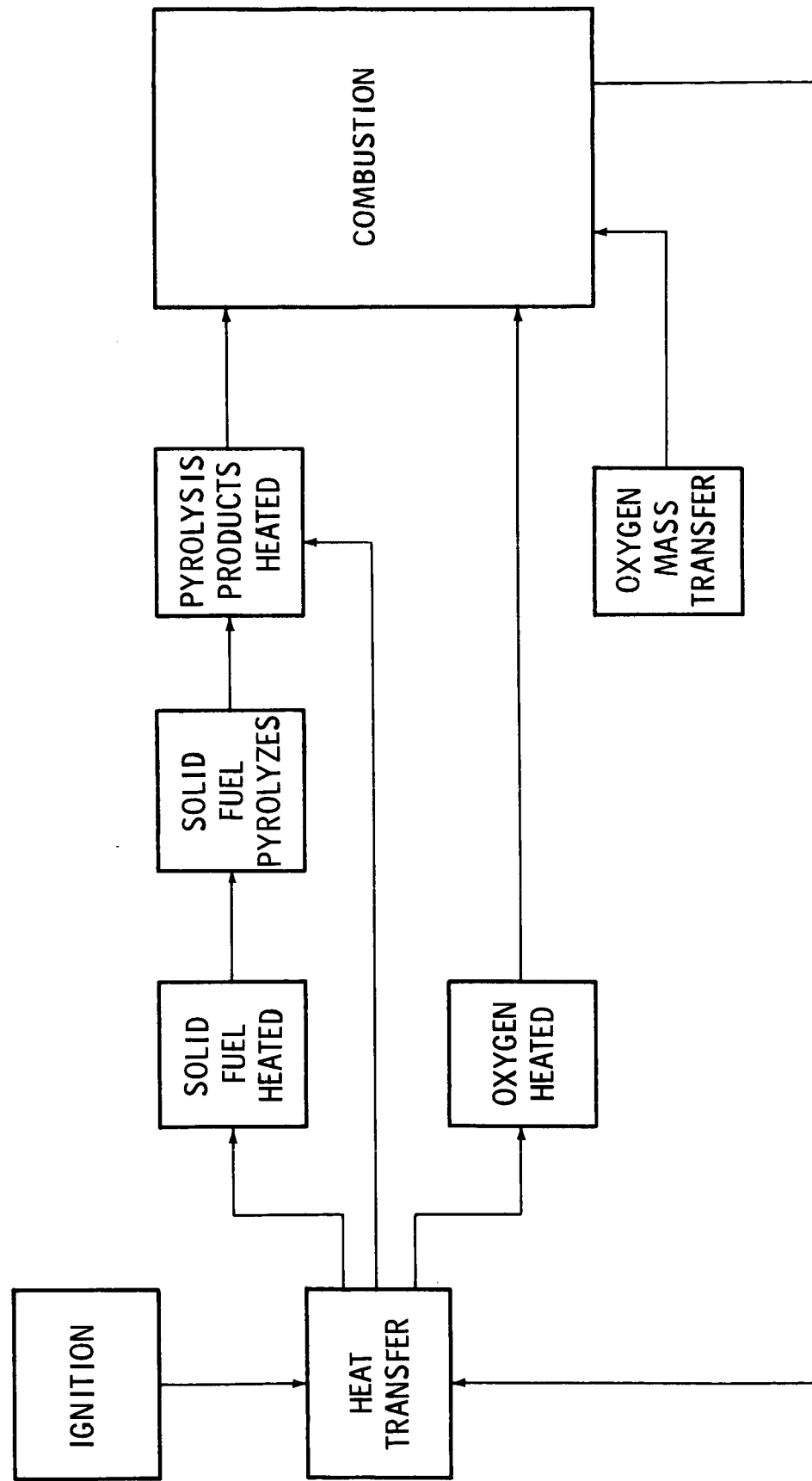


Fig. 10 - Flow of Causation for Combustion of Solids.

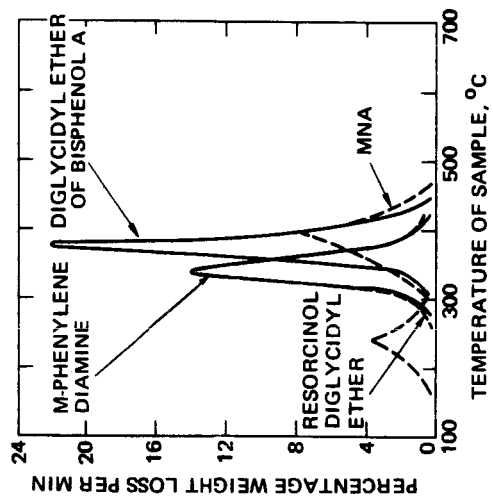


Fig. 11 - Differential Thermogravimetric Curves for Epoxide Polymers; Heating Rate 5° c/min. (From Reich and Levi¹⁹).

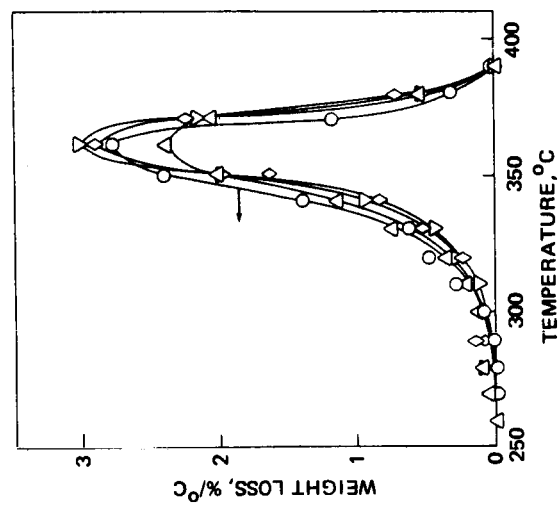


Fig. 12 - Thermogravimetric Analysis Curves
for Styrene-Acrylonitrile Copolymers
of different Molecular Weight. (O) 1.2×10^4 ;
(Δ) 4.6×10^4 ; (O) 18.4×10^4 ; (∇) 73.6×10^4 (66).
(From Reich and Levi²⁰).

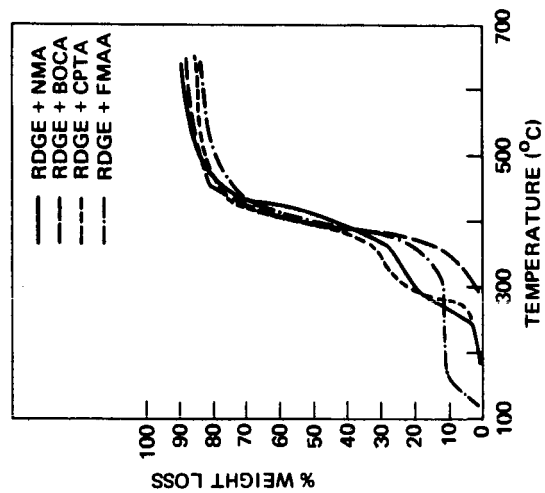


Fig. 13 - Thermograms of Resorcinol Diglycidyl Ether Cured with Several Diels - Alder Adducts.

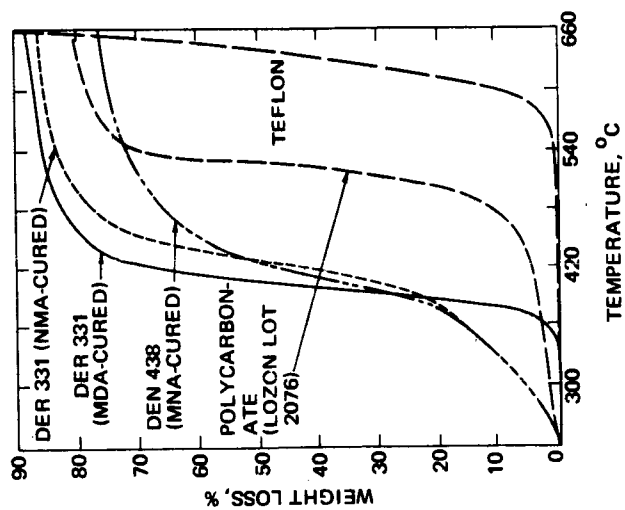


Fig. 14 - Thermogravimetric Curves of Phenolic Condensation Polymers; Heating Rate 9 - 10° c/min.

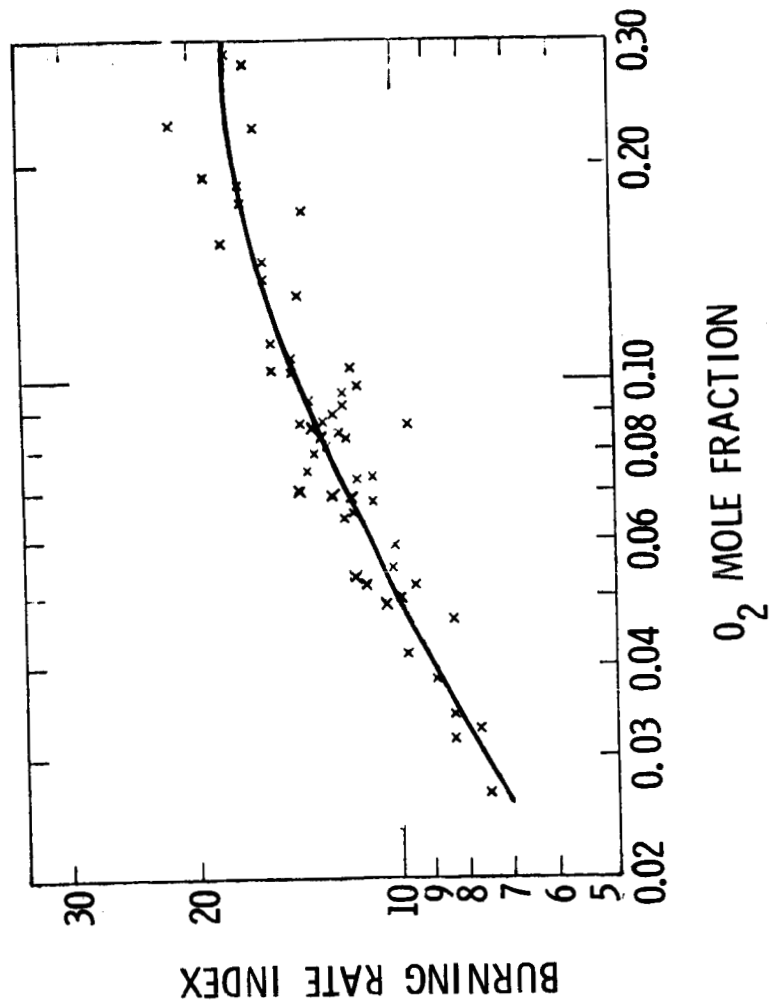


Fig. 15 - Variation of Burning Rate of Propane with Mole Fraction of Oxygen. (From Hottel et al.²⁴).

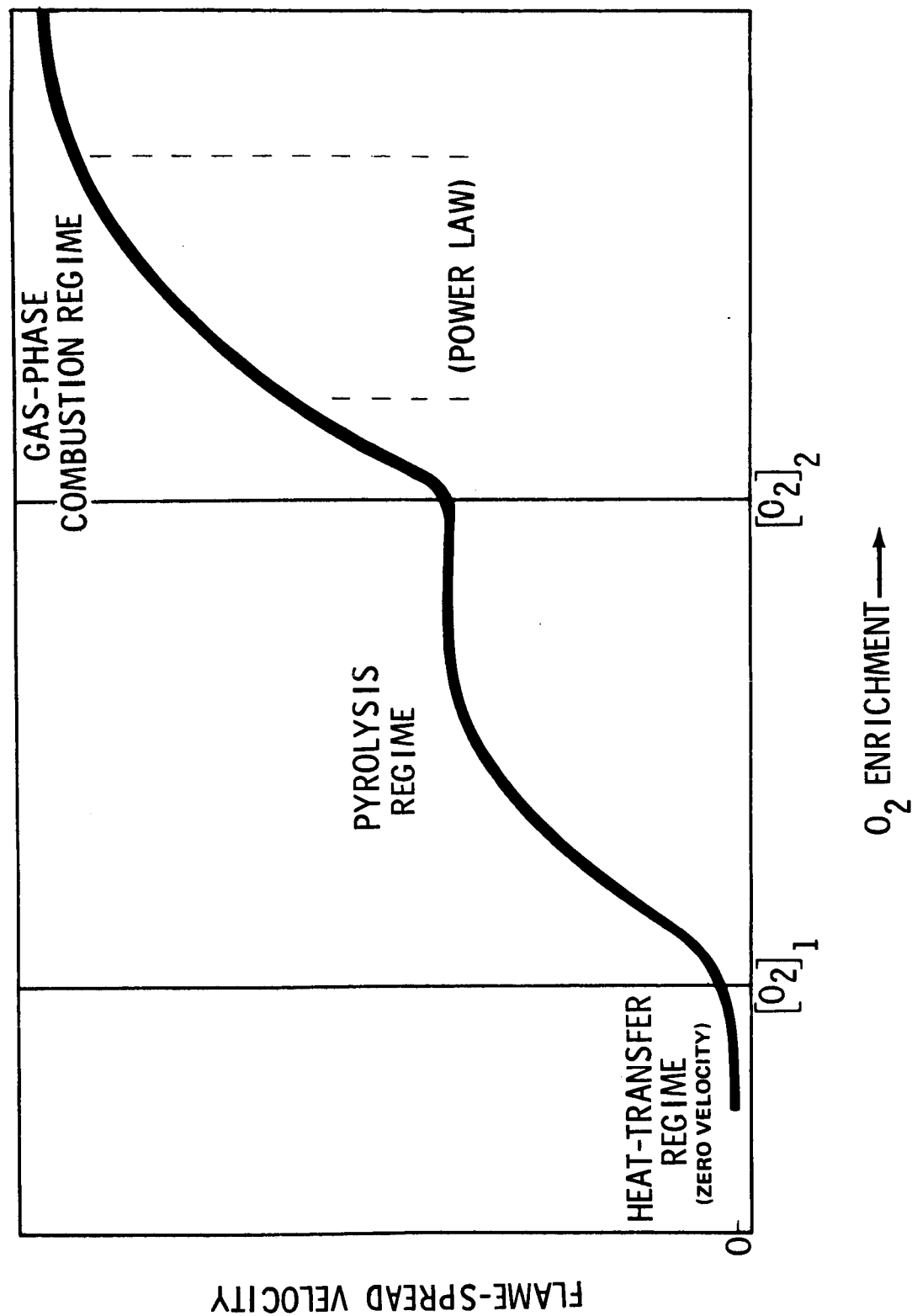


Fig. 16 - Typical Flame Spread Velocity Versus O_2 Enrichment Curve as Interpreted by Hypothesis.